

Aircraft Observations of
Extreme Ozone Concentrations
Near Thunderstorms

(U.S.) Environmental Protection Agency
Research Triangle Park, NC

1985



U.S. Department of Commerce
National Technical Information Service

NTIS

TECHNICAL REPORT DATA		
(Please read Instructions on the reverse before completing)		
1. REPORT NO. EPA/600/J-85/504	2.	3. PB88-139837
4. TITLE AND SUBTITLE AIRCRAFT OBSERVATIONS OF EXTREME OZONE CONCENTRATIONS NEAR THUNDERSTORMS (Journal Version)	5. REPORT DATE 1985	
	6. PERFORMING ORGANIZATION CODE	
7. AUTHOR(S) John F. Clarke and George W. Griffing	8. PERFORMING ORGANIZATION REPORT NO.	
9. PERFORMING ORGANIZATION NAME AND ADDRESS (same as Block 12)	10. PROGRAM ELEMENT NO. CDWA1A/02-1336(FY-84)	
	11. CONTRACT/GRANT NO.	
12. SPONSORING AGENCY NAME AND ADDRESS Atmospheric Sciences Research Laboratory OFFICE OF RESEARCH AND DEVELOPMENT U.S. ENVIRONMENTAL PROTECTION AGENCY RESEARCH TRIANGLE PARK, NORTH CAROLINA 27711	13. TYPE OF REPORT AND PERIOD COVERED	
	14. SPONSORING AGENCY CODE EPA/600/09	
15. SUPPLEMENTARY NOTES Atmospheric Environment, 19(7):1175-1179, July 1985		
16. ABSTRACT Anomalously large short-term ozone concentrations were observed on several occasions by aircraft during an experiment on August 5, 1980, to characterize the physical and chemical properties of the Baltimore urban plume. The ozone "spikes" of about 500 ppb were traversed by aircraft in less than 30 s (travel distance of less than 2 km). Analysis of these and ancillary data suggest that the ozone spikes may have resulted from ozone production by chemical reactions activated by lightning associated with thunderstorms.		
17. KEY WORDS AND DOCUMENT ANALYSIS		
a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
REPRODUCED BY U.S. DEPARTMENT OF COMMERCE NATIONAL TECHNICAL INFORMATION SERVICE SPRINGFIELD, VA. 22161		
18. DISTRIBUTION STATEMENT RELEASE TO PUBLIC	19. SECURITY CLASS (This Report) UNCLASSIFIED	21. NO. OF PAGES 8
	20. SECURITY CLASS (This page) UNCLASSIFIED	22. PRICE PC89.95 / MF 695

AIRCRAFT OBSERVATIONS OF EXTREME OZONE CONCENTRATIONS NEAR THUNDERSTORMS

JOHN F. CLARKE* and GEORGE W. GRIFFING*

Meteorology and Assessment Division, Environmental Sciences Research Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, U.S.A.

(First received 10 July 1984 and in final form 18 December 1984)

Abstract—Anomalous large short-term ozone concentrations were observed on several occasions by aircraft during an experiment on 5 August 1980 to characterize the physical and chemical properties of the Baltimore urban plume. The ozone 'spikes' of about 500 ppb were traversed by aircraft in less than 30 s (travel distance of less than 2 km). Analysis of these and ancillary data suggest that the ozone spikes may have resulted from ozone production by chemical reactions activated by lightning associated with thunderstorms.

Key word index: Ozone, thunderstorms, field study, lightning, nitrogen oxides.

1. INTRODUCTION

During July and August 1980, the U.S. Environmental Protection Agency sponsored the Northeast Regional Oxidant Study (NEROS) to obtain data for developing and evaluating a regional-scale transport and dispersion model for ozone (O_3) and O_3 precursors (Clarke and Ching, 1983; Clark and Clarke, 1984). As part of this study, the Baltimore, MD, urban plume was extensively sampled for O_3 and other chemical and meteorological properties for 20 days. The measurements were taken at fixed locations, with a mobile van, and by two aircraft. On one of the days, 5 August 1980, two spike-like O_3 concentrations were observed during aircraft traversals downwind of Baltimore. The largest spike exceeded 500 ppb in a background concentration of less than 200 ppb and lasted less than 30 s (about 2 km of aircraft travel). These data are presented and discussed.

2. BACKGROUND

On 5 August a warm front extended from Connecticut through the Great Lakes. Hot, humid, hazy weather conditions prevailed in the eastern half of the country. Cirrus clouds were present over much of the area with cumulonimbus clouds developing in the area by early afternoon. By mid-afternoon thunderstorms occurred in the Washington-Baltimore area. The boundary layer flow was generally westerly at $3-5 \text{ ms}^{-1}$ during the morning, becoming southwesterly by early afternoon. The Baltimore urban plume extended to the northeast and was extensively sampled by aircraft during the afternoon. The location

of the ground-level projections of the aircraft traversals and the starting and ending times of the traversals are shown in Fig. 1. The aircraft were operated by Brookhaven National Laboratory (BNL) (dashed lines) and Washington State University (WSU) (solid lines). Also shown in the figure is the location of the BNL instrumented van 25 km northeast of downtown Baltimore and the locations of the spike-like O_3 occurrences 17 km north of Baltimore and 58 km northeast of Baltimore.

Ozone concentrations measured at the instrumented van were approximately 60 ppb at about 1000 EST, increasing to a maximum of nearly 200 ppb at 1245 EST. The air above the mixed layer (from aircraft spirals) had an O_3 concentration of about 80 ppb. Analysis of maximum 1-h concentrations from surface sites suggests a broad plume emanating from the Washington-Baltimore complex and a large area with concentrations exceeding 180 ppb downwind of Baltimore. Maximum O_3 concentrations were generally observed prior to 1400 EST.

3. OZONE SPIKES

3.1. First example

Time plots of relative humidity, scattering coefficient (B_{scat}), O_3 concentration and nitrogen dioxide (NO_2) concentration for measurements during a WSU flight between 1422 and 1436 EST (roughly equivalent to 40 km) are shown in Fig. 2. The measurements were taken crosswind 15 km downwind from the center of Baltimore (see Fig. 1) at an altitude of about 754 m. A spike-like anomaly on the O_3 concentration trace occurred between 1425 and 1426 EST. The spike maximum was 553 ppb, with a width at half maximum of about 1 km. This spike and others discussed here were checked against the original aircraft strip chart and accepted as real. The actual magnitude of the spike was probably considerably larger than the reported

*On assignment from the National Oceanic and Atmospheric Administration, U.S. Department of Commerce.

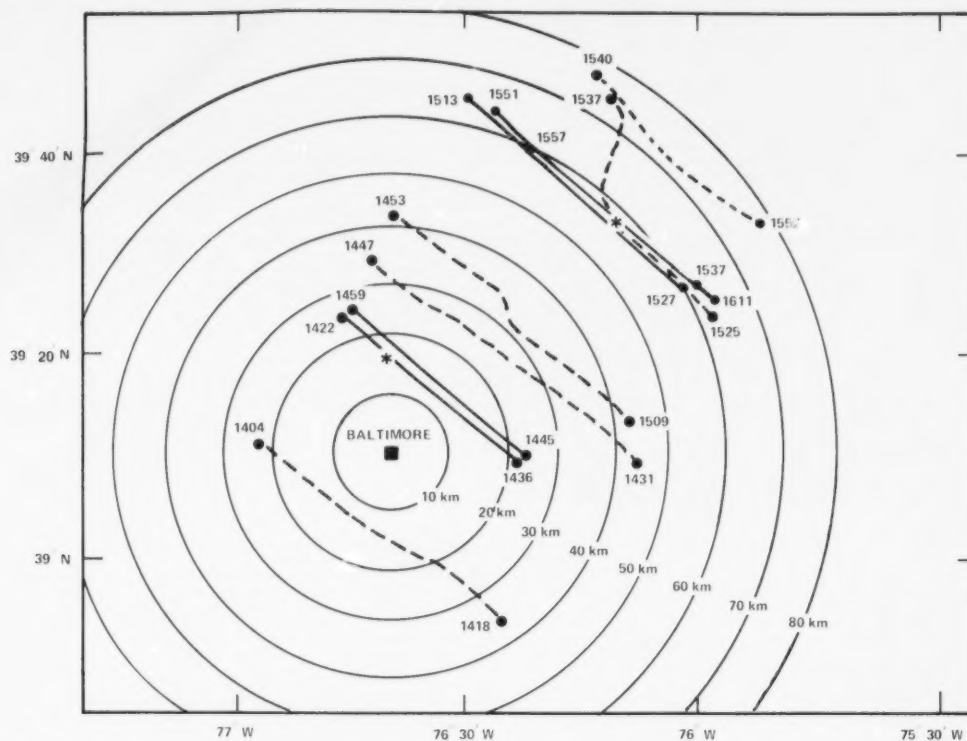


Fig. 1. Aircraft trajectories for WSU (solid) and BNL (broken) with respect to the center of Baltimore. Beginning and ending times are indicated (EST). The asterisks indicate the locations of the spike-like O_3 concentration features. The open circle indicates the location of the van.

553 ppb considering the response time of the instrument and the averaging period of the data. Characteristics of the O_3 and nitrogen oxide/oxides of nitrogen (NO/NO_x) instruments flown aboard the WSU and BNL aircraft are shown in Table 1.

After completing the measurements shown in Fig. 2, the WSU aircraft flew in the opposite direction at an altitude of 1300 m. The ozone spike was not observed on this latter traverse. This is not inconsistent with the

reality of the spike in Fig. 2, since the relatively small (lateral extent) anomaly would have been advected almost 10 km downwind and dispersed considerably in the 30 min that elapsed between the measurements.

3.2. Second example

Time plots of relative humidity, scattering coefficient, O_3 concentration and NO_2 concentration for measurements during a WSU aircraft flight between

Table 1. Instrument characteristics

	WSU	BNL
Ozone		
Instrument	CSI 2000	AID560
Analysis technique	Chemiluminescent	Chemiluminescent
Resolution	5 ppb	5 ppb
Response time	5 s	5 s
Averaging time	11 s	4 s
NO/NO_x		
Instrument	ML 8440 HP	TECO 14b
Analysis technique	Chemiluminescent	Chemiluminescent
Resolution	5 ppb	6 ppb
Response time	5 s	11 s
Averaging time	11 s	4 s
Location accuracy	6 km	6 km

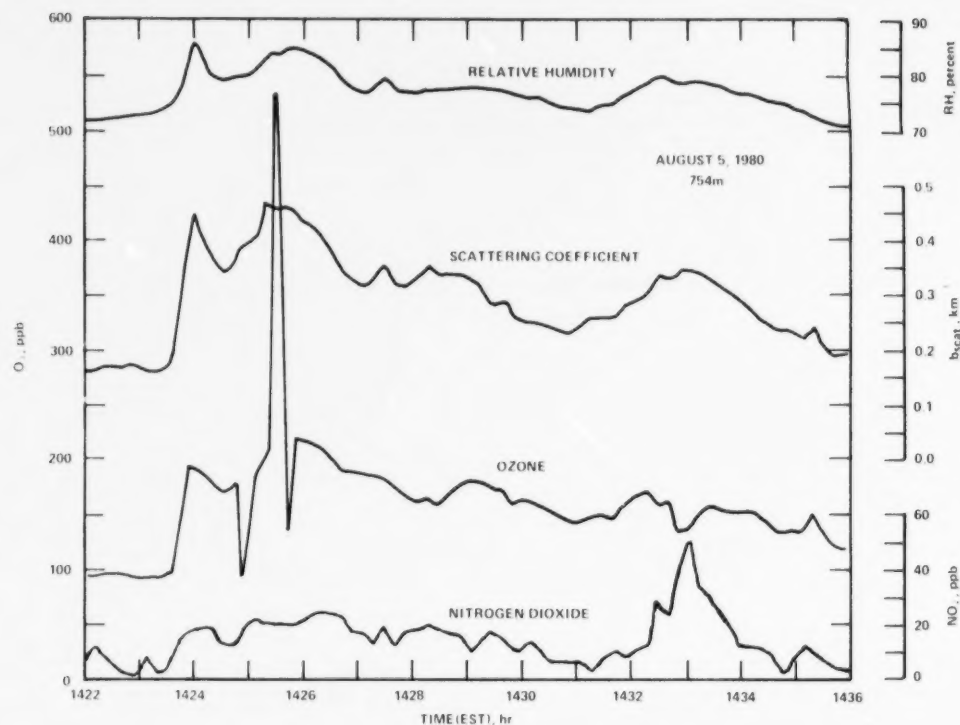


Fig. 2. Traces of measurements by the WSU aircraft at an altitude of 754 m and 15 km downwind of Baltimore.

1513 and 1527 EST are shown in Fig. 3. The measurements were taken crosswind about 60 km downwind of Baltimore at an altitude of 456 m. A spike-like anomaly on the O_3 concentration trace occurred at 1521 EST. It had a maximum of about 457 ppb and a width at half maximum of about 1 km. For this example the BNL observations provide corroborating evidence of the reality of the anomaly.

Measurements with the BNL aircraft were taken at near the same time and same location at an altitude of 433 m (see Fig. 1). Time plots of the BNL measurements between 1525 and 1537 EST are shown in Fig. 4. There is a prominent anomaly on the O_3 concentration trace between 1531 and 1533 EST. The arrow indicates the position of the O_3 spike observed at 1521 EST by WSU. The O_3 concentration anomaly observed by BNL was at approximately the same position as the O_3 spike observed by WSU 10 min earlier. Advection over that period of time could transport the O_3 spike 3–5 km which, for the location accuracy of the platforms (see Table 1) is consistent with the similarity of positions. Diffusion, in the time interval between the aircraft observations, would be expected to broaden the spike and reduce the peak concentration, as observed. The spatial proximity and similarity of the O_3 concentration anomalies observed by BNL and WSU suggest that they are real and of common origin.

4. DISCUSSION

Tropospheric O_3 concentrations are a composite of contributions from three different mechanisms (Vassily, 1965): (1) chemical reactions of natural and anthropogenic precursor emissions, (2) exchange of tropospheric and stratospheric atmospheres and (3) chemical reactions activated by anthropogenic and natural electrical discharges. The first mechanism is probably the major source of tropospheric O_3 during the summer months. However, it is extremely unlikely that the O_3 spikes in Figs 2, 3 and 4 resulted from this mechanism. For the prevailing convective meteorological conditions, the source of a plume with the width of the spikes would be located only about 10 km or 30 min travel time upwind, hardly sufficient time to produce O_3 concentrations of the magnitude observed. In addition, the B_{scat} and NO_2 traces do not exhibit characteristics to be expected from a strong coherent point source plume. Also, if such a source had existed, it would have been observed during other aircraft transects in the July–August sampling period.

We thus turn our attention to the second and third mechanisms. Tropospheric and stratospheric atmospheres may be exchanged through thunderstorm activity, which also produces electrical discharges. Increases of the O_3 concentration at ground level

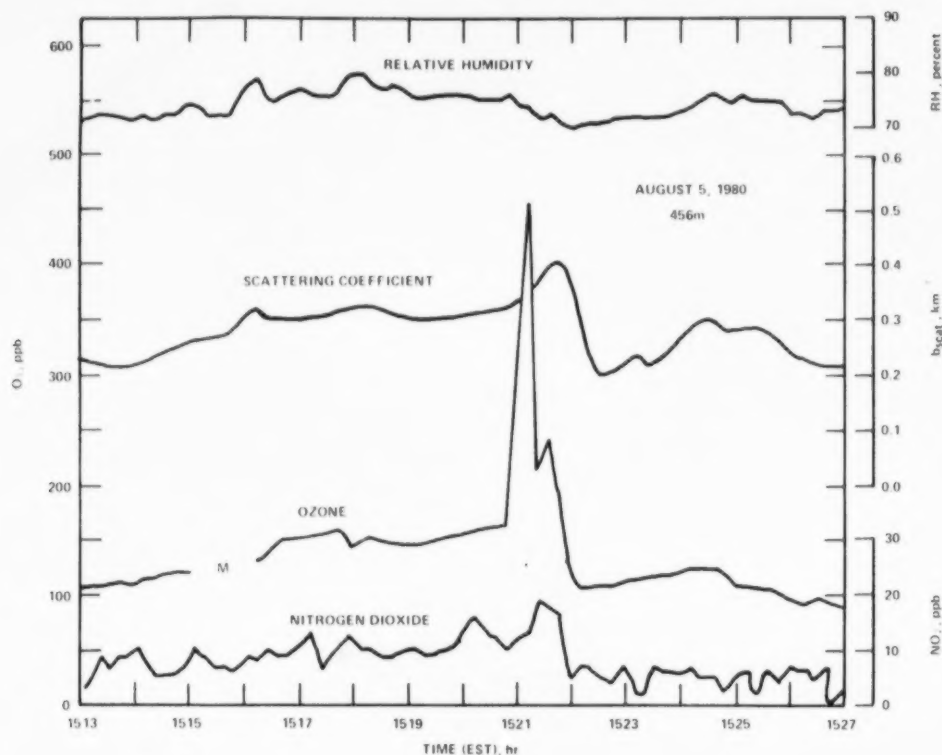


Fig. 3. Traces of measurements by the WSU aircraft at an altitude of 456 m and 60 km downwind of Baltimore.

during thunderstorms have been observed by Vassy (1965) and Attmannspacher and Hartmannsgruber (1973). Using spectroscopic techniques, Orville (1967) analyzed the spectrum of a lightning flash and concluded that O_3 concentration along a light path of about 0.85 km was an order of magnitude larger than the ambient O_3 . Salanave (1970) used a spectrograph and u.v. source to study the increase of O_3 concentration after a thunderstorm. He found increases of O_3 concentration at ground level almost a factor of 3 of the O_3 concentration before the thunderstorm.

Shlanta and Moore (1972) studied the distribution of O_3 associated with New Mexico thunderstorms. Transient increases of the O_3 concentration from a background of about 30 ppb to a maximum of about 200 ppb were observed to occur with lightning-induced emissions of negative charge from an exposed and grounded metal rod. The duration of the transients appeared to be in the order of a second.

Thunderstorms were occurring in the aircraft sampling area on the afternoon of 5 August 1980. A satellite photograph taken at 1431 EST indicated thunderstorms in the vicinity of where the first O_3 protuberance was observed. A notation on the recorder strip chart in the WSU aircraft indicated that the aircraft was flying through rain at 1411 EST. An earlier notation indicated that the aircraft was flying

near cumulus clouds. A radar summary at 1535 EST indicated numerous echos over the area with tops to almost 17 km. Thus, the mechanisms existed for O_3 to be either imported from the stratosphere or produced locally by chemical processes associated with lightning discharges.

Thunderstorm downdrafts from the stratosphere, when they extend to near the surface, are usually larger than the observed diameter of the spikes and carry with them clean, dry air characteristic of the stratosphere. This was not the case with either of the spikes. Thus, we favor the explanation that the ozone spikes were produced locally by chemical processes associated with lightning discharges.

According to theory (Griffing, 1977), the production of NO_2 would also be expected from chemical reactions activated by electrical discharges associated with thunderstorms. Solar absorption techniques were used by Noxon (1976) to show that NO_2 was produced during a thunderstorm. More recently, the production of NO_2 by lightning in a thunderstorm was observed by Drapeho *et al.* (1983). The NO_2 concentration traces in Figs 2-4 do not suggest the same extreme chemical activity associated with the production of the ozone spikes. However, for the second example (Figs 3 and 4) there is correlation of O_3 and NO_2 in the vicinity of the spikes and NO_2 concentrations are in

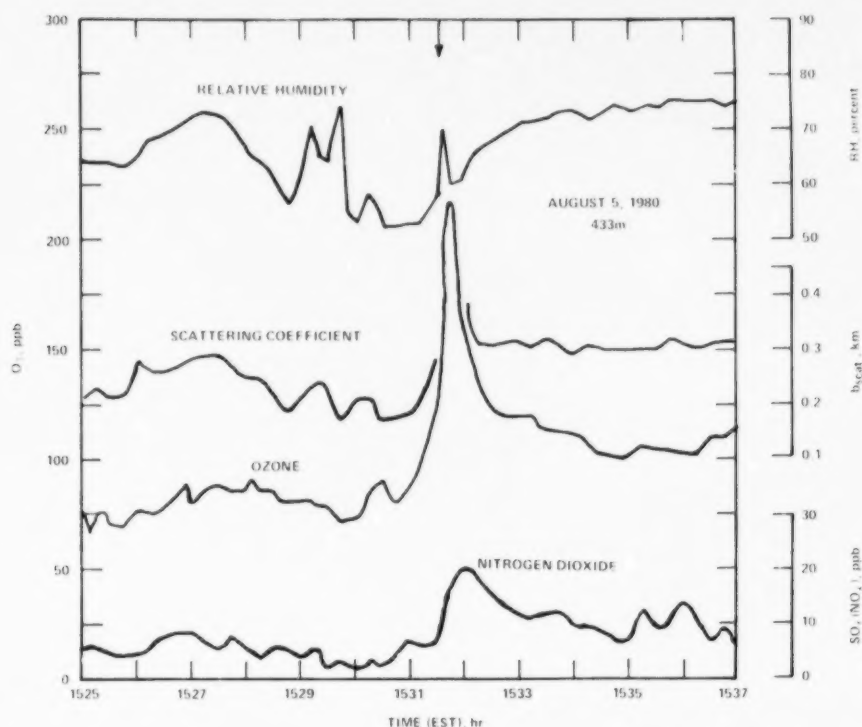


Fig. 4. Traces of measurements by the BNL aircraft at an altitude of 433 m and 65 km downwind of Baltimore. The arrow indicates the corresponding location of the spike-like ozone concentration feature of Fig. 3.

agreement with those reported by Drapcho *et al.* (1983) as due to lightning discharges.

5. CONCLUSIONS

Unusual spike-like O_3 concentrations were observed on two occasions downwind of Baltimore on 5 August 1980. Review of ancillary data including the backup aircraft strip chart indicates that the O_3 spikes represent valid measurements and were associated with thunderstorm activity. We suggest the spikes were a result of chemical activity initiated by lightning discharges. If this premise is correct, numerous O_3 spikes probably occurred on the afternoon of 5 August that went undetected both within and above the boundary layer. Most of the spikes could be quickly dispersed in the convective systems. However, they could make a contribution to the O_3 burden. Documenting these cases is therefore important for a fuller understanding of all processes relating to the O_3 budget of the boundary layer.

REFERENCES

- Attmannspacher W. and Hartmannsgruber R. (1973) On extremely high values of ozone near the ground. *Pure appl. Geophys.* **106-108**, 1091-1096.
- Clarke J. F. and Ching J. K. S. (1983) Aircraft observations of regional transport of oxidant in the northeastern United States. *Atmospheric Environment* **17**, 1703-1712.
- Clark T. L. and Clarke J. F. (1984) A Lagrangian study of the boundary layer transport of pollutants in the northeastern United States. *Atmospheric Environment* **18**, 287-297.
- Drapcho D. L., Sisterson D. and Kumar R. (1983) Nitrogen fixation by lightning activity in a thunderstorm. *Atmospheric Environment* **17**, 729-734.
- Griffing G. W. (1977) Ozone and oxides of nitrogen production during thunderstorms. *J. geophys. Res.* **82**, 943-950.
- Noxon J. F. (1976) Atmospheric nitrogen fixation by lightning. *Geophys. Res. Lett.* **3**, 463-465.
- Orville R. E. (1967) Ozone production during thunderstorms, measured by ultraviolet radiation from lightning. *J. geophys. Res.* **72**, 3557-3561.
- Salanave L. E. (1970) Ozone absorption in spectra of lightning and an ultraviolet source (abstract). *Eos. Trans. AGU* **5**, 757.
- Shlanta A. and Moore C. B. (1972) Ozone and point discharge measurements under thunderstorms. *J. geophys. Res.* **77**, 4500-4510.
- Vassy A. (1965) Atmospheric ozone. *Adv. Geophys.* **11**, 115-173.

NOTICE

This document has been reviewed in accordance with U.S. Environmental Protection Agency policy and approved for publication. Mention of trade names or commercial products does not constitute endorsement or recommendation for use.

This article documents research that was conducted under the auspices of the U.S. Environmental Protection Agency (USEPA). As an arm of the government, the USEPA has a paid up, non-exclusive, irrevocable, worldwide license to reproduce, prepare derivative works, and distribute to the public all of the material contained herein, and to grant others acting on its behalf the right to do so. The USEPA grants to the National Technical Information Service, Department of Commerce, the right to act as its agent in the distribution of this material.